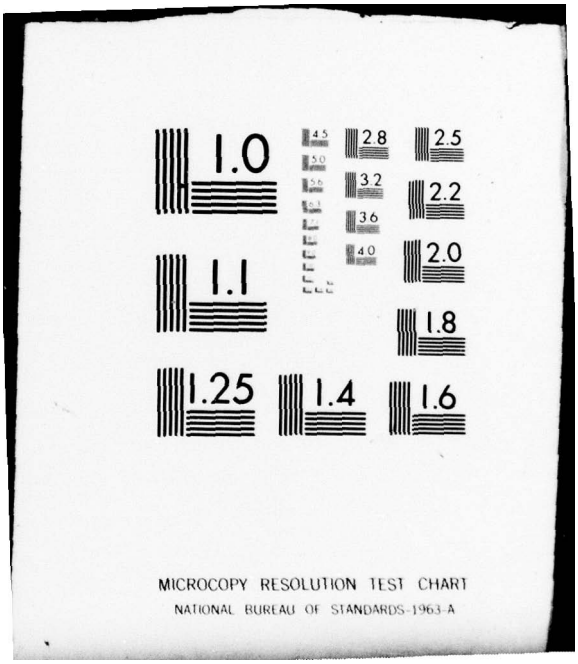
AD-A071 853 COLORADO UNIV BOULDER F/G 20/5
OPTICAL AND DISCHARGE STUDIES OF NOVEL ELECTRONIC TRANSITION LA--ETC(U)
JUN 79 S R LEONE N00014-77-C-0115
UNCLASSIFIED NL

| OF |
AD A071853

END
DATE FILMED
8-79
DDC


1.0
1.1
1.25
1.4
1.6
1.8
2.0
2.2
2.5
2.8
3.2
3.6
4.0
MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

NR 395-571
Code 421

12 B.S.

# LEVEL III

(9) INTERIM TECHNICAL REPORT, (1 June 1979)

ONR Contract: N00014-77-C-0115

A059979



(15) N00014-77-C-0115

(10) Principal Investigator: Stephen R. Leone

Institution: University of Colorado
Boulder, Colorado 80309

(6) Title: "Optical and Discharge Studies of Novel Electronic Transition Laser Species"

ARPA Order No: 2840 Amend. 12

Contract Date: 78 Dec. 01

Contract Amount: $40,000

(11) 1 Jun 79 (12) 6p.

Accomplishments:

A meter long, transverse, high temperature discharge laser device was successfully constructed. The device consists of a quartz laser tube, with 10 pairs of tantalum wire electrodes, each 10 cm long, spaced 2 cm apart. The tantalum electrodes were gold-brazed to tungsten feedthroughs which were fused directly into the quartz with GS-1 glass. Ten 2.7 nF capacitors on each side of the laser tube formed a voltage doubling circuit with approximately 15 Joules of energy storage. The experimentally determined discharge parameters were: 50 kV with a 15 nsec current risetime which was measured optically. These characteristics were highly effective in producing the usual 3371 Å laser transition in nitrogen, with 3 mJ of energy.

The entire laser tube was housed in a high temperature oven, consisting of 120° sector ceramic potted heaters and hollowed out firebricks. The 15 nsec risetime achieved was excellent, considering the long current return paths necessitated by the bulky oven. The laser tube itself sustains 800°C operation under high vacuum ($10^{-6}$ Torr). The tungsten feedthroughs are maintained at a 250°C lower temperature than the main body of the tube in order to reduce corrosion rates with the hot sulfur vapor.

Introduction of sulfur vapor into the laser tube was found to be complicated by several problems. Contact of the hot sulfur vapor with the tantalum and tungsten electrode materials generated several new species, one of which is volatile at room temperature, and another which has significant vapor pressure at high temperatures. Even though gross macroscopic decomposition of the electrode materials was very slow, the generation of these new gases was rapid each time the sulfur was introduced

into the tube. These new materials had two serious effects. The high temperature compound absorbs in the visible, appearing to be an extremely strong, broad-banded absorber. The brilliant blue discharges observed in test cells of $S_2$ vapor were modified to give only weaker intensity red or yellow discharges. There were several differences in our test cell designs from the final laser tube. In one test cell, a longitudinal discharge was used, so that the electrodes were always maintained at low temperatures (200°C) while the main length of $S_2$ vapor was at 700°C. In the second test cell, a transverse geometry was used, with solid tantalum bar stock for electrodes and gold plating on stainless steel feedthroughs. In both of these cells, the generation of new compounds was not observed on heating, nor were the reddish-yellow discharges. Intense blue discharges were always observed from these test cells. There were several different features in the meter long laser, primarily with regard to the tungsten feedthroughs, which were required to make reliable seals, and the use of tantalum wire electrodes.

The observation of severe material problems points out how serious the incompatibility is for the corrosive $S_2$ vapor. It appears that there will really be no metal materials fully compatible with sulfur vapor at the temperatures and pressures required.

It is conceivable, but unlikely, that the problem of the strong visible absorber is linked to other species in the sulfur vapor composition. For example, $S_4$ is known to absorb in the visible. Normally, however, at low vapor pressures ($\sim$few torr) and high temperatures (600-700°C), the concentration of $S_4$ is very small and has never posed a problem in all quartz cells. The behavior of the visible absorbing compound is not at all characteristic of what would occur if it were $S_4$. Thus, it is likely that the visible absorber is some metal sulfide, which becomes more volatile at higher temperatures.

Because of the continual presence of the strong visible absorber at the temperatures needed, it is highly unlikely that stimulated emission could be observed in $S_2$. The changeover of the brilliant blue discharges to dim yellow-red discharges further substantiates the presence of the absorbing vapor at the required temperatures and pressures and their deleterious effect on the chances for lasing.

Summary of Conclusions from the Discharge $S_2^{\pi}$ Laser Work:

(1) Electric discharges in pure $S_2^{\pi}$ vapor do produce brilliant blue glow discharges characteristic of the $S_2^{\pi}$(B-X) emission.

(2) Sulfur vapor is extremely corrosive and is effectively incompatible with all metal materials at 600°C.

Prognosis for the Future:

So far, none of the physics involved would indicate that an electric discharge $S_2$ laser is not possible. Other schemes might be employed to generate $S_2$ at lower temperatures. For example, a variety of mercury, cadmium, etc., sulfides decompose to give pure $S_2$ species. It might be possible to generate $S_2$ in an all quartz region and flow the vapor through the metal electrodes zone at low temperatures (200°C). However, this would prevent a sealed-off, continuous use system from being developed, and involves a great deal of additional technological experimental effort. In our opinion, such further effort is not prudent at this time.

Other Efforts:

Several other experimental efforts are ongoing and the results of these works have been prepared for publication. A cumulative list of relevant ONR sponsored publications is:

1. S. R. Leone and K. G. Kosnik, "A Tunable Visible and Ultraviolet Laser on $S_2(B^3\Sigma_u^- - X^3\Sigma_g^-$," Appl. Phys. Lett. 30, 346 (1977).

2. J. B. Koffend, R. W. Field, D. R. Guyer, and S. R. Leone, "Pulsed and cw Optically Pumped Lasers for Novel Applications in Spectroscopy and Kinetics," Laser Spectroscopy, Proceedings of the Third International Conference on Laser Spectroscopy, Jackson Hole, Wyoming, J. L. Hall and J. L. Carlsten, Ed. Springer Verlag, Springer Series in Optical Sciences (1977), Vol. 7, p. 382.

3. H. Hofmann and S. R. Leone, "Collisional Deactivation of Laser-Excited $Br^*(^2P_{1/2})$ Atoms with Halogen and Interhalogen Molecules," Chem. Phys. Lett. 54, 314 (1978).

4. H. Hofmann and S. R. Leone, "Quenching and Reactions of Laser-Excited $I(5^2P_{1/2})$ Atoms with Halogen and Interhalogen Molecules," J. Chem. Phys. 69, 641 (1978).

5. H. Hofmann and S. R. Leone, "Tunable Laser Photodissociation of $HgI_2$: Quantum Yield for Formation of Excited $I(5^2P_{1/2})$ Atoms," J. Chem. Phys. 69, 3819 (1978).

6. S. L. Baughcum, H. Hofmann, S. R. Leone and D. J. Nesbitt, "Photofragmentation Dynamics and Reactive Collisions of Laser-Excited Electronic States," to be published, Faraday Discussions, 67 (1979).

In addition, the principal investigator has been asked to deliver invited talks on the subject of optically pumped electronic transition lasers at two important meetings: The Gordon Conference on Lasers and Nonlinear Optics (Wolfeboro, NH), and The XI International Conference on the Physics of Electron and Atomic Collisions (Kyoto, Japan).

**15.5 Cascade Lasing and Collisional Excitation Transfer in Optically Pumped Calcium Atoms.**[1] W. H. Pence and S. R. Leone, *Joint Institute for Laboratory Astrophysics, National Bureau of Standards and University of Colorado and Department of Chemistry, University of Colorado, Boulder, CO 80309*

*(15 min)*

Stimulated emission is observed in atomic calcium at 6717 Å, 6102 Å, 6122 Å, 6162 Å, and 1.9853 μm, while optically pumping the $4^1S_0 \rightarrow 5^1P_1$ transition at 2721.6 Å. All of the transitions except 6717 Å arise through a rapid collisional excitation transfer from the $5^1P_1$ to the $5^3P_{2,1,0}$ levels, resulting in cascade lasing, $5^3P_2 \rightarrow 5^3S_1 \rightarrow 4^3P_{2,1,0}$. The stimulated emission at 6717 Å on the $5^1P_1 \rightarrow 3^1D_2$ transition and the multiplet $5^3S_1 \rightarrow 4^3P_{2,1,0}$ at 6162 Å, 6122 Å, and 6102 Å exhibit the characteristic shortened pulse of a "bottlenecked" laser due to the metastability of the lower-laser levels. The $5^3P_2 \rightarrow 5^3S_1$ transition at 1.9853 μm exhibits no bottleneck and the stimulated emission follows precisely the time development of the 2 μsec optical pumping pulses.

The apparatus consists of a flashlamp-pumped dye laser operating on Coumarin 504, frequency doubled to provide the optical pump at 2721.6 Å. An etalon is used to narrow the laser line width to ~0.3 $cm^{-1}$. A cell constructed of quartz with a tantalum liner is filled with a few grams of Ca metal, pumped out to $10^{-6}$ torr, and backfilled with 0 to 25 torr of argon gas for the collisional transfer measurements. The cell is equipped with brewster angle windows and two fluorescence ports. The tantalum-lined portion of the cell is maintained at 600°C in an oven. Laser emission is monitored with photodiodes and a 77°K Ge:Au infrared detector. Visible fluorescence is observed with photomultiplier tubes and narrow bandpass interference filters to isolate single-line emission. The kinetics of the energy transfer process are followed by monitoring the ratio of intensities of fluorescence lines in the singlet and triplet manifolds as experimental parameters are varied. The signals from the lines are observed simultaneously and signal averaged using a transient digitizer and a signal averager.

Upon optical excitation at 2721.6 Å, stimulated emission at 6717 Å ($5^1P_1 \rightarrow 3^1D_2$) is observed at calcium pressures between 1 millitorr and 10 torr. The $3^1D_2$ level is metastable and consequently the population inversion is rapidly terminated, resulting in a laser pulse of ~50 nsec in duration. The pumping pulse from the dye laser is ~2 μsec. Emission from the lower-laser level at 4575 Å ($3^1D_2 \rightarrow 4^1S_0$) occurs on a time scale too short to accurately measure with the long pumping pulse, but we estimate its lifetime to be on the order of 600 nsec.

Collisional transfer efficiently and rapidly populates the $5^3P_{2,1,0}$ levels in the triplet manifold. Very strong stimulated emission is observed from the $3^3P_2$ level to the $5^3S_1$ level. Since the lower-laser level here is not bottlenecked, the 1.9853-μm emission follows the pumping pulse exactly. The time to threshold for this emission is 30 to 50 nsec. The $5^3S_1$ level, populated by this stimulated emission, also lases at 6162 Å, 6122 Å, and 6102 Å to the metastable $4^3P_{2,1,0}$ levels. The intensities appear approximately in the statistical weights of the lines. This transition is bottlenecked and the laser pulses have a duration of ~50 nsec.

The $5^1P_1 \rightarrow 5^3P_{2,1,0}$ energy transfer occurs with both $Ca^* + Ca$ and $Ca^* + Ar$ collisions. By monitoring a ratio of fluorescence intensities of emission from lines in both the singlet and triplet manifolds, a determination of the cross sections for collisional transfer can be made. Preliminary measurements on the collisional transfer with argon were made by holding the calcium vapor pressure to ~10 millitorr. At this pressure the contribution to the $5^1P_1 \rightarrow 5^3P_{2,1,0}$ transfer by $Ca^* + Ca$ is negligible. Upon varying the argon pressure from 0.3 to 15 torr we find that the ratio of triplet to singlet fluorescence intensities increases smoothly by a factor of 3, reaching a plateau. The data fits the expected dependence on argon pressure for a simple equilibrium electronic-to-electronic energy transfer. It is evident that the transfer cross sections are very large, indicating an efficient resonant energy transfer between the $5^1P_1$ level and the $5^3P_{2,1,0}$ manifold. More accurate data which will provide specific cross sections for the collisional excitation transfer will be presented at the conference.

[1]This work was sponsored by the Office of Naval Research.